

Description

10/516918

Process and apparatus for reducing the content of NO<sub>x</sub> and N<sub>2</sub>O in gases

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The present invention relates to a process for reducing the content of nitrogen oxides in gases, in particular in process gases and offgases, and also an apparatus suitable for carrying out this process.

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Many processes, e.g. combustion processes or in the industrial preparation of nitric acid, produce an off-gas laden with nitrogen monoxide NO, nitrogen dioxide NO<sub>2</sub> (together referred to as NO<sub>x</sub>) and nitrous oxide N<sub>2</sub>O.

15 While NO and NO<sub>2</sub> have long been known as compounds having ecotoxic relevance (acid rain, smog formation) and limit values for maximum permissible emissions of these have been set worldwide, nitrous oxide has increasingly moved into the focus of environmental  
20 protection in recent years, since it contributes to a not inconsiderable extent to the degradation of stratospheric ozone and to the greenhouse effect. For reasons of environmental protection, there is an urgent need for technical solutions which enable nitrous oxide  
25 emissions to be eliminated together with the NO<sub>x</sub> emissions.

Numerous possible ways of separately removing firstly N<sub>2</sub>O and secondly [lacuna] are known.

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In the case of NO<sub>x</sub> reduction, particular mention may be made of the selective catalytic reduction (SCR) of NO<sub>x</sub> by means of ammonia in the presence of vanadium-containing TiO<sub>2</sub> catalysts (cf., for example, G. Ertl, H. Knözinger J. Weitkamp: Handbook of Heterogeneous Catalysis, Vol. 4, pages 1633-1668, VCH Weinheim (1997)). This can, depending on the catalyst, proceed at temperatures of from about 150°C to about 450°C and makes it possible to decrease the concentration of NO<sub>x</sub>

by more than 90%. It is the most widely utilized variant of the removal of  $\text{NO}_x$  from offgases from industrial processes.

- 5 Processes for the reduction of  $\text{NO}_x$  which are based on zeolite catalysts and proceed using a variety of reducing agents are also known. Apart from Cu-exchanged zeolites (cf., for example, EP-A-0914866), iron-containing zeolites in particular appear to be of  
10 interest for practical use.

Thus, US-A-4,571,329 claims a process for reducing  $\text{NO}_x$  in a gas comprising at least 50% of  $\text{NO}_2$  by means of ammonia in the presence of an Fe zeolite. The ratio of  
15  $\text{NH}_3$  to  $\text{NO}_2$  is at least 1.3. According to the process described in this patent,  $\text{NO}_x$ -containing gases are said to be reduced by means of ammonia without formation of  $\text{N}_2\text{O}$  as by-product occurring.

- 20 US 5,451,387 describes a process for the selective catalytic reduction of  $\text{NO}_x$  by means of  $\text{NH}_3$  over iron-exchanged zeolites at temperatures of about 400°C.

In contrast to the removal of  $\text{NO}_x$  from offgases, which  
25 has been established industrially for many years, there are only few industrial processes for eliminating  $\text{N}_2\text{O}$ . These are usually based on thermal or catalytic degradation of the  $\text{N}_2\text{O}$ . An overview of catalysts whose in-principle suitability for the degradation and  
30 removal of nitrous oxide has been demonstrated, is given by Kapteijn et al. (Kapteijn F. et al., Appl. Cat. B: Environmental 9 (1966) 25-64).

Fe and Cu zeolite catalysts which either effect a pure  
35 decomposition of the  $\text{N}_2\text{O}$  into  $\text{N}_2$  and  $\text{O}_2$  (US-A-5,171,553) or catalytically reduce the  $\text{N}_2\text{O}$  with the aid of  $\text{NH}_3$  or hydrocarbons to  $\text{N}_2$  and  $\text{H}_2\text{O}$  or  $\text{CO}_2$  appear to be particularly useful.

Thus, JP-A-07 060 126 describes a process for reducing  $N_2O$  by means of  $NH_3$  in the presence of iron-containing zeolites of the pentasil type at temperatures of  $450^\circ C$ . The reduction in the  $N_2O$  concentration which can be  
5 achieved by means of this process is 71%.

In Catal. Lett. 62 (1999) 41-44, Mauvezin et al. give an overview of the suitability of various, iron-exchanged zeolites of the MOR, MFI, BEA, FER, FAU, MAZ  
10 and OFF types for this purpose. According to this reference, reduction of more than 90% of the  $N_2O$  can be achieved by addition of  $NH_3$  at below  $500^\circ C$  only in the case of Fe-BEA.

15 Apart from the abovementioned processes for the separate removal of  $N_2O$  and  $NO_x$ , there are also processes for combined removal which can be carried out using a single catalyst.

20 WO-A-00/48715 discloses a process in which an  $NO_x$ - and  $N_2O$ -containing offgas is passed at temperatures of from  $200$  to  $600^\circ C$  over an iron zeolite catalyst of the beta type (= BEA type), with the offgas additionally containing  $NH_3$  in a ratio of from  $0.7$  to  $1.4$  based on  
25 the total amount of  $NO_x$  and  $N_2O$ . Here,  $NH_3$  serves as reducing agent both for  $NO_x$  and for  $N_2O$ . Although the process operates at temperatures of less than  $500^\circ C$ , it suffers, like the abovementioned process, from the in-principle disadvantage that an approximately equimolar  
30 amount of reducing agent (here  $NH_3$ ) is required to eliminate the  $N_2O$  content.

WO-A-01/51,181 discloses a process for the removal of  $NO_x$  and  $N_2O$ , in which a process gas or offgas is passed  
35 through two reaction zones containing iron-laden zeolites as catalysts. In the first reaction zone,  $N_2O$  is removed, ammonia is added to the gas mixture between the first and second reaction zones and  $NO_x$  is reduced in the second reduction zone.

It has now surprisingly been found that the effectiveness of the abovementioned process can be increased significantly when the reduction in the  $N_2O$  content to the desired degree of removal occurs not solely in the first reaction zone, but instead the reaction zone for the  $NO_x$  reduction can also be utilized for reducing the  $N_2O$  content. This has become possible since it has surprisingly been established that simultaneous  $NO_x$  reduction (e.g. by means of  $NH_3$ ) and  $N_2O$  decomposition is possible when using iron-laden zeolite catalysts. The contribution of the second reaction step to the decomposition of  $N_2O$  is particularly great when the process is operated at elevated pressures, i.e. at pressures above 2 bar, preferably above 4 bar.

It is an object of the present invention to provide a simple but economical process which gives good conversions both for  $NO_x$  removal and for  $N_2O$  removal and has minimal operating and capital costs. The former includes not only the energy for setting the necessary operating temperature but also the consumption of reducing agent and energy losses due to resistances to flow in the catalyst bed (pressure drops). The capital costs are determined essentially by the required amounts of catalyst and the apparatus volumes associated therewith.

In addition, there is the problem of introducing the reducing agent which has to be mixed intimately with the gas stream to be treated in order to ensure a very high efficiency of the reducing agent (avoidance of secondary reactions and passage without reaction). The mixer necessary for this purpose should take up as little space as possible because of installation and economic considerations.

These objects are achieved by the process of the

invention and the apparatus of the invention.

The invention provides a process for reducing the content of  $\text{NO}_x$  and  $\text{N}_2\text{O}$  in gases, in particular in process gases and offgases, which comprises the measures:

- 10 a) passing the  $\text{N}_2\text{O}$ - and  $\text{NO}_x$ -containing gas over a sequence of two catalyst beds comprising one or more iron-laden zeolites,
- b) adding a reducing agent for  $\text{NO}_x$  between the catalyst beds,
- c) setting a temperature of less than  $500^\circ\text{C}$  in the first catalyst bed and second catalyst bed,
- 15 d) setting a gas pressure of at least 2 bar in the two catalyst beds,
- e) selecting a space velocity in the first and second catalyst beds such that a reduction in the  $\text{N}_2\text{O}$  content of the gas by not more than 90%, based on the  $\text{N}_2\text{O}$  content at the entrance to the first catalyst bed, occurs in the first catalyst bed and that a further reduction in the  $\text{N}_2\text{O}$  content of the gas by at least 30%, based on the  $\text{N}_2\text{O}$  content at the entrance to the second catalyst bed, occurs in the second catalyst bed.

In the first catalyst bed for pure  $\text{N}_2\text{O}$  decomposition, the  $\text{NO}_x$  which is still present in the gas accelerates, as expected, the desired  $\text{N}_2\text{O}$  decomposition by means of an activating action as has been described for various  $\text{N}_2\text{O}/\text{NO}_x$  ratios by Kögel et al. in Catal. Comm. 2 (2001) 273-6.

35 However, an appreciable decrease in the  $\text{N}_2\text{O}$  concentration by decomposition into nitrogen and oxygen can be achieved in the second catalyst bed, too. This was surprising since, firstly, the  $\text{NO}_x$  content which activates the  $\text{N}_2\text{O}$  decomposition is reduced by addition

of the reducing agent and, secondly, it was expected that the reducing agent added would be temporarily adsorbed on the catalyst surface and thus block the active sites for  $N_2O$  decomposition.

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Under the chosen process conditions, i.e. the elevated pressures and in particular a reduced  $NH_3/NO_x$  ratio, these influences obviously do not come to bear.

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The process of the invention thus makes it possible to carry out both the decomposition of  $N_2O$  and the reduction of  $NO_x$  at a low operating temperature and economical space velocities and at the same time achieve high degrees of removal of  $N_2O$  and  $NO_x$ .

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For the purposes of the present invention, the space velocity is the volume of gas mixture (measured at  $0^\circ C$  and 1.014 bara) per hour divided by the volume of catalyst. The space velocity can thus be set via the volume flow of the gas and/or via the amount of catalyst.

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The gas laden with nitrogen oxides is usually passed over the catalyst at a space velocity of from 200 to 200,000  $h^{-1}$ , preferably from 5,000 to 100,000  $h^{-1}$ , in particular from 5,000 to 50,000  $h^{-1}$ , based on the total catalyst volume of the two catalyst beds.

25

After leaving the first catalyst bed, the  $N_2O$  content is, according to the process of the invention, preferably above 200 ppm, in particular above 300 ppm. A reduction of not more than 90%, preferably not more than 80%, in the  $N_2O$  content present at the beginning of the first catalyst bed occurs in the first catalyst bed.

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After leaving the first catalyst bed, the  $N_2O$ - and  $NO_x$ -containing gas is firstly mixed with a gaseous reducing agent, preferably with  $NH_3$ , and subsequently passed

over the catalyst at a temperature of preferably less than 450°C and at the chosen space velocity to achieve simultaneous removal of N<sub>2</sub>O (by decomposition) and NO<sub>x</sub> (by reduction).

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In the second catalyst bed, an additional reduction of at least 30%, preferably at least 40%, in the N<sub>2</sub>O content present at the beginning of the second catalyst bed occurs.

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In the process of the invention, iron-containing zeolites are used in the first and second catalyst beds. The catalysts used in the respective catalyst beds can be different or can preferably be the same catalyst.

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Physical separation of the catalyst beds makes it possible to set the temperature of the second catalyst bed or of the gas stream entering it by removal or introduction of heat so that it is lower or higher than that of the first catalyst bed.

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The temperature of the gas stream in the first catalyst bed in which only the N<sub>2</sub>O is removed and also in the second catalyst bed in which N<sub>2</sub>O and NO<sub>x</sub> are removed is, according to the invention, below 500°C, preferably in the range from 250 to 500°C, in particular from 300 to 450°C and very particularly preferably from 350 to 450°C. The temperature in the second catalyst bed preferably corresponds to the temperature in the first catalyst bed. The temperature in the catalyst bed can advantageously be determined as the arithmetic mean of the temperature of the gas stream at the entrance to and exit from the catalyst bed.

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The choice of operating temperature is, like the space velocity selected, determined by the desired degree of removal of N<sub>2</sub>O.

The temperature, volume flow and amount of catalyst in the first catalyst bed are preferably selected so that not more than 90%, preferably not more than 80% and very particularly preferably not more than 70%, of the N<sub>2</sub>O present at the beginning of the first catalyst bed is decomposed in the first catalyst bed.

The temperature, volume flow and amount of catalyst in the second catalyst bed are preferably selected so that a further reduction in the N<sub>2</sub>O content of the gas by at least 30%, based on the N<sub>2</sub>O content at the entrance to the second catalyst bed, occurs in the second catalyst bed.

The process of the invention is carried out at an elevated pressure of at least 2 bar, preferably at least 3 bar, very particularly preferably from 4 to 25 bar. The introduction of the reducing agent between the first catalyst bed and the second catalyst bed, i.e. downstream of the first and upstream of the second catalyst bed, is carried out by means of a suitable device, e.g. an appropriate pressure valve or appropriately configured nozzles.

In the first reaction zone, a relatively low water concentration is generally preferred, since a very high water content would make high operating temperatures (e.g. >500°C) necessary. Depending on the type of zeolite used and the time of operation, this could exceed the hydrothermal stability limits of the catalyst. However, the NO<sub>x</sub> content plays a critical role here, since this can stop deactivation by water.

In the case of the NO<sub>x</sub> reduction in the second reaction zone, a high water content plays a minor role, since high degrees of removal of NO<sub>x</sub> are achieved here even at relatively low temperatures.

The reducing agent is used in the amount required for



reduction of the  $\text{NO}_x$ . For the purposes of the present description, this is that amount of reducing agent which is necessary to reduce the  $\text{NO}_x$  present in the gas mixture either completely or to the desired final  
5 concentration without appreciable reduction of  $\text{N}_2\text{O}$  taking place.

As reducing agents for the purposes of the invention, it is possible to use substances which have a high  
10 activity and selectivity for the reduction of  $\text{N}_2\text{O}$  and whose selectivity and activity under the reaction conditions selected is greater than that for the possible reduction of  $\text{N}_2\text{O}$ .

15 Reducing agents which can be used for the purposes of the invention are, for example, hydrocarbons, hydrogen, carbon monoxide, ammonia or mixtures thereof, e.g. synthesis gas. Particular preference is given to ammonia or substances which liberate ammonia when they  
20 are introduced, e.g. urea or ammonium carbamate.

The amount of reducing agent added must not be appreciably greater than that required for the reduction of  $\text{NO}_x$  under the reaction conditions  
25 selected.

In the case of ammonia as reducing agent, use is made, depending on the desired degree to which the  $\text{NO}_x$  content is to be decreased, of up to a maximum of 1.2, preferably from 1.0 to 1.2, mol of ammonia per mol of  
30  $\text{NO}_x$ . If a relatively low degree of removal of  $\text{NO}_x$  is desired, the maximum amount of ammonia is  $1.2 \cdot y$  mol per mol of  $\text{NO}_x$ ;  $y$  is the percentage of  $\text{NO}_x$  which is to be consumed in the reduction.

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When suitable catalysts and process conditions are chosen, the  $\text{NH}_3$  added does not act as reducing agent for  $\text{N}_2\text{O}$  but instead selectively reduces the  $\text{NO}_x$  present in the offgas.

The process of the invention thus makes it possible to carry out the removal of  $N_2O$  and of  $NO_x$  at a lower operating temperature with a small consumption of gaseous reducing agent, e.g.  $NH_3$ , which has hitherto not been possible by means of the processes described in the prior art.

This is a particularly great advantage when large amounts of  $N_2O$  are to be eliminated.

The way in which the gaseous reducing agent is introduced into the gas stream to be treated can be chosen freely for the purposes of the invention as long as the introduction is upstream of the second catalyst bed. The reducing agent can be introduced, for example, in the inlet line upstream of the container for the second catalyst bed or immediately before the catalyst bed. The reducing agent can be introduced in the form of a gas or else a liquid or aqueous solution which vaporizes in the gas stream to be treated.

Catalysts used according to the invention substantially comprise one or more iron-laden zeolites, preferably in an amount of >50% by weight, in particular >70% by weight. Thus, for example, not only an Fe-ZSM-5 zeolite but also a further iron-containing zeolite, e.g. an iron-containing zeolite of the MFI or FER type, can be present in the catalyst used according to the invention. In addition, the catalyst used according to the invention can further comprise other additives known to those skilled in the art, e.g. binders.

Catalysts used according to the invention are preferably based on zeolites into which iron has been introduced by solid-state ion exchange. The commercially available ammonium zeolites (e.g.  $NH_4$ -ZSM-5) and appropriate iron salts (e.g.  $FeSO_4 \times 7 H_2O$ ) are usually used as starting materials for this purpose and

are intensively mixed with one another by mechanical means in a ball mill at room temperature. (Turek et al.; Appl. Catal. 184, (1999) 249-256; EP-A-0 955 080). The references are hereby expressly incorporated by  
5 reference. The catalyst powders obtained are subsequently calcined in air at temperatures in the range from 400 to 600°C in a muffle furnace. After calcination, the iron-containing zeolites are intensively washed in distilled water, filtered off and  
10 dried. The iron-containing zeolites obtained in this way are subsequently admixed with suitable binders and mixed and, for example, extruded to form cylindrical catalyst bodies. Suitable binders include all binders customarily used; the most widely used here are  
15 aluminum silicates such as kaolin.

According to the present invention, the zeolites which can be used are laden with iron. The iron content can be up to 25%, but is preferably from 0.1 to 10%, based  
20 on the mass of zeolite.

Iron-laden zeolites are preferably of the MFI, BEA, FER, MOR, FAU and/or MEL type, in particular of the ZSM-5 type.  
25

In a preferred embodiment, iron-laden zeolites whose crystal structure has no pores or channels having crystallographic diameters greater than or equal to 7.0 Ångstrom are used at least in the second catalyst bed.  
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These include iron-laden zeolites of the MFI, FER and/or MEL type, in particular of the ZSM-5 type.

The process of the invention can also be carried out  
35 using zeolites in which part of the lattice aluminum has been isomorphously replaced by one or more elements, for example by one or more elements selected from among B, Be, Ga, Fe, Cr, V, As, Sb and Bi. The use of zeolites in which the lattice silicon is

isomorphously replaced by one or more elements, for example by one or more elements selected from among Ge, Ti, Zr and Hf, is likewise included.

- 5 Precise details on the configuration or structure of the zeolites used according to the invention are given in the Atlas of Zeolite Structure Types, Elsevier, 4th revised edition 1996, which is hereby expressly incorporated by reference.

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In the process of the invention, very particular preference is given to using the above-defined zeolite catalysts which have been treated with water vapor (steamed catalysts). Such treatment removes aluminum from the lattice of the zeolite, and is known per se to those skilled in the art. These hydrothermally treated zeolite catalysts surprisingly display a particularly high activity in the process of the invention.

- 20 Preference is given to using hydrothermally treated zeolite catalysts which have been laden with iron and in which the ratio of extralattice aluminum to lattice aluminum is at least 1:2, preferably from 1:2 to 20:1.

- 25 The water content of the reaction gas is preferably in the range <25% by volume, in particular in the range <15% by volume. A low water content is generally to be preferred.

- 30 In general, a relatively low water concentration is preferred, since relatively high water contents would make relatively high operating temperatures. This could, depending on the type of zeolite used and the operating time, exceed the hydrothermal stability limits of the catalyst and is thus to be matched to the particular case chosen.
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The presence of CO<sub>2</sub> and of other deactivating constituents of the reaction gas which are known to

those skilled in the art should also be minimized if possible, since they would have an adverse effect on the N<sub>2</sub>O removal.

5 The process of the invention also works in the presence of O<sub>2</sub>, since the catalysts used according to the invention have appropriate selectivities which suppress reaction of the gaseous reducing agent, e.g. NH<sub>3</sub>, with O<sub>2</sub> at temperatures of <500°C.

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All these influencing factors and also the chosen throughput over the catalyst, i.e. the space velocity, are to be taken into account in choosing the appropriate operating temperature of the reaction zone.

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The process of the invention can be used in particular in nitric acid production, for offgases from power stations or for gas turbines. Process gases and offgases which contain nitrogen oxides and from which  
20 the nitrogen oxides can be removed inexpensively by means of the process disclosed here are obtained in these processes. The process of the invention is advantageously used for the tailgas from nitric acid production downstream of the absorption tower.

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The configuration of the catalyst beds can be chosen freely for the purposes of the invention. Thus, for example, the catalyst or catalysts can be arranged in a catalyst bed through which the gas flows axially or  
30 preferably radially in one or more containers.

The invention further provides an apparatus for reducing the content of NO<sub>x</sub> and N<sub>2</sub>O in gases, in particular in process gases and offgases, which  
35 comprises:

- A) two catalyst beds which are connected in series and each comprise one or more iron-laden zeolites and through which the NO<sub>x</sub>- and N<sub>2</sub>O-

containing gas flows,

B) a device for introducing a gaseous reducing agent into the stream of the  $\text{NO}_x$ - and  $\text{N}_2\text{O}$ -containing gas located between the catalyst beds, wherein

C) the  $\text{NO}_x$ - and  $\text{N}_2\text{O}$ -containing gas flows radially through at least one of the catalyst beds.

In a preferred embodiment, both catalyst beds are arranged in one container which considerably reduces the apparatus costs.

According to the invention, the gas to be purified flows radially through at least one catalyst bed, preferably both catalyst beds, which results in a considerably reduced pressure drop.

The catalyst beds through which the gas flows radially are, for example, configured in the form of hollow cylinders but can also have other shapes. The catalyst beds through which the gas flows radially can be arranged above one another or a combination of catalyst beds through which axial and radial flow occurs can be chosen. The path of the gas is prescribed by suitably arranged separators between the catalyst beds so that the gas flows firstly through the first catalyst bed and then through the second catalyst bed.

In the case of catalyst beds through which the gas flows radially, these can also be in the form of hollow cylinders arranged concentrically within one another. In this embodiment too, it should be ensured that the path of the gas is prescribed by suitably arranged separators between the catalyst beds so that the gas flows firstly through the first catalyst bed and then through the second catalyst bed.

The flow direction of the gas in the radial screen reactor can be from the inside outward or from the

outside inward.

In a preferred embodiment, there are two catalyst beds through which the gas flows radially, for example in the form of two hollow cylinders, having different dimensions, with the external dimension of one catalyst bed being smaller than the internal dimension of the other catalyst bed and both catalyst beds being arranged concentrically, and with the path of the gas being prescribed by suitably arranged separators between the catalyst beds so that the gas flows firstly through the first catalyst bed and then through the second catalyst bed.

In a further preferred embodiment of the apparatus of the invention, the gas which has passed through the first catalyst bed is passed into a mixer which is preferably located in the center of the apparatus and a feed line for reducing agent which opens into the space downstream of the first catalyst bed and before or preferably into the mixer is provided, with the gas to be purified being passed through the second catalyst bed after leaving the mixer.

The mixer serves to distribute the reducing agent intimately in the gas stream. For the purposes of the invention, the mixer can be configured freely, for example as a static mixer with appropriate internals or as a dynamic mixer. The simplest form of a tube through which flow is preferably turbulent is also possible as a mixer for the purposes of the invention.

Figures 1 to 6 show preferred embodiments of the apparatus of the invention in longitudinal section.

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Figure 1 depicts an apparatus according to the invention with gas inlet (1) and gas outlet (2). In the upper interior space closest to the gas inlet (1), the first catalyst bed is arranged in the form of a hollow

cylinder (4) and is located on a dividing wall which divides the interior of the apparatus into two halves. In addition, the upper end face of the hollow cylinder (4) is closed by a dividing wall. The gas to be purified flows through the gas inlet (1) and, via the annular gap of the entrance (7) to the first catalyst bed, flows radially through the first catalyst bed into the annular gap of the exit (8) from the first catalyst bed. From there it flows into the mixer (6) at whose inlet end there is the opening of an inlet line (3) for the reducing agent. Mixer (6) passes through the dividing wall and the gas then flows, via the annular gap of the entrance (9) to the second catalyst bed (5) located beneath the first catalyst bed (4), radially through the second catalyst bed into the annular gap of the exit (10) from the second catalyst bed (5). From there, the purified gas leaves the apparatus via the gas outlet (2).

Figure 2 shows a similar embodiment as Figure 1 with the differences that the first catalyst bed (4) is located underneath the second catalyst bed (5) and that gas inlet (1) and gas outlet (2) are arranged laterally in the apparatus. The other references numerals have the meanings given in the description of Figure 1.

Figure 3 depicts a further embodiment of the apparatus of the invention with gas inlet (1) and gas outlet (2). Here, the first catalyst bed (4) and the second catalyst bed (5) are configured as two hollow cylinders arranged concentrically within one another. The first catalyst bed (4) is located outside a concentric dividing wall (11) which closes off the lower end face of the catalyst bed (4), the annular gaps (7) and (8) and also the interior of the apparatus and the upper end face of the second catalyst bed (5). The gas to be purified enters the apparatus through the gas inlet (1) and flows radially through the first catalyst bed from the outer annular gap entrance (7) inward to the



annular gap exit (8). From there it flows into the mixer (6) at whose inlet end there is the opening of an inlet line (3) for the reducing agent. The mixer (6) opens into the interior of the second catalyst bed (5) which is closed off at the bottom by a dividing wall. The gas then flows, via the annular gap of the entrance (9) to the second catalyst bed (5), radially outward through the second catalyst bed into the annular gap of the exit (10) from the second catalyst bed (5). From there the purified gas leaves the apparatus via the gas outlet (2).

Figure 4 shows a similar embodiment as Figure 3 with the difference that the first catalyst bed (4) forms the inner hollow cylinder and the second catalyst bed (5) forms the outer hollow cylinder. The other reference numerals have the meanings given in the description of Figure 3.

Figure 5 shows an embodiment in which one catalyst bed through which the gas flows axially and one catalyst bed through which the gas flows radially are provided. The gas flows via the gas inlet (1) axially through the first catalyst bed (4) and into the mixer (6). The apparatus has a dividing wall which divides the interior of the apparatus into two halves. At the inlet end of the mixer (6) there is the opening of an inlet line (3) for the reducing agent. From the mixer (6) the gas flows into the annular gap of the entrance (9) to the second catalyst bed (5) and radially through this into the annular gap of the exit (10). From there the purified gas leaves the apparatus via the gas outlet (2).

Figure 6 shows a similar embodiment as Figure 5 with the difference that the gas flows radially through the first catalyst bed (4) and axially through the second catalyst bed (5). The other reference numerals have the meanings given in the description of Figure 3.

The process of the invention is illustrated by the following example.

5 The catalyst used was an iron-laden zeolite of the ZSM-5 type. The Fe-ZSM-5 catalyst was prepared by solid-state ion exchange starting from a commercially available zeolite in ammonium form (ALSI-PENTA, SM27). Details regarding the preparation can be taken from:  
10 M. Rauscher, K. Kesore, R. Mönig, W. Schwieger, A. Tißler, T. Turek: "Preparation of highly active Fe-ZSM-5 catalyst through solid state ion exchange for the catalytic decomposition of  $N_2O$ " in Appl. Catal. 184 (1999) 249-256.

15 The catalyst powders were calcined in air at 823 K for 6 hours, washed and dried overnight at 383 K. After addition of appropriate binders, the powders were extruded to form cylindrical catalyst bodies.

20 As apparatus for reducing the  $NO_x$  and  $N_2O$  content, use was made of two tube reactors which were connected in series and were each charged with such an amount of the above catalyst that a space velocity of  $15,000\text{ h}^{-1}$  based  
25 on the inflowing gas stream resulted in each case.  $NH_3$  gas was added between the two reaction zones. The operating temperature of the reaction zones was set by means of heating. The analysis of the gas streams entering and leaving the reactors was carried out by  
30 means of an FTIR gas analyzer.

At inlet concentrations of 1,500 ppm of  $N_2O$ , 350 ppm of  $NO_x$ , 3,000 ppm of  $H_2O$  and 1.2% by volume of  $O_2$  and  $N_2$  and with intermediate addition of  $NH_3$  at a uniform  
35 operating temperature of  $425^\circ\text{C}$  and an operating pressure of 6.5 bar, the conversion results for  $N_2O$ ,  $NO_x$  and  $NH_3$  listed in the following table were obtained.

Table

	Inlet concentration	Outlet concentration	Conversion
N <sub>2</sub> O	1,500 ppm (reactor 1)	540 ppm (reactor 1)	64%
NO <sub>x</sub> (x=1-2)	360 ppm (reactor 2)	80 ppm (reactor 2)	78%
NH <sub>3</sub>	310 ppm*) (reactor 2)	0 ppm (reactor 2)	100%
N <sub>2</sub> O	540 ppm (reactor 2)	190 ppm (reactor 2)	65%

\*) added between the first reactor and the second reactor